Renewable and Sustainable Energy Reviews 16 (2012) 4638–4650

Contents lists available at SciVerse ScienceDirect

Renewable and Sustainable Energy Reviews

journal homepage: www.elsevier.com/locate/rser

Environmental life cycle assessment of lignocellulosic conversion to ethanol: A review

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ARTICLE INFO

Article history:
Received 12 January 2012
Received in revised form
3 April 2012
Accepted 6 April 2012
Available online 27 June 2012

Keywords:
LCA
Lignocelluloses
Bioethanol
Environmental impact
Sustainability

ABSTRACT

Bioenergy from lignocellulosic biomass offers the potential to provide a significant source of clean, low carbon and secure energy. In recent years, a number of studies have been carried out to assess the environmental performance of lignocellulosic ethanol fuel. However, the complexity of biofuel systems generates significantly different results due to the differences in input data, methodologies applied, and local geographical conditions. Moreover, much attention has been placed on assessing climate change potential and energy consumption. This study draws on 53 published life cycle assessment of the lignocellulosic ethanol. More than half of the articles reviewed focus on assessing greenhouse gas (GHG) emission or fossil energy consumption or combination of both. All studies but two reviewed conclude that there is a reduction of GHG emission when using lignocellulosic ethanol in comparison to fossil fuel reference system. However, different studies have reported different sources contributing to GHG emission: some reports majority of GHG emissions come from biomass cultivation stage; others argue significant GHG emissions from ethanol conversion process. All articles suggest a reduction of fossil consumption in all cases of ethanol fuel. Contrary results for the impact of acidification and eutrophication potential from lignocellulosic ethanol are also observed—some reports less impact in comparison to conventional gasoline whiles others report significant increase of acidification and eutrophication potential by ethanol production. Studies also show water consumption varies significantly depending on biomass types, irrigation requirement, and regional irrigation practices; with different findings on whether agricultural practices or ethanol conversion being the main sources for water consumption. Contrary findings on emissions contributing to ecotocixity and human health have also been reported with some being favourable while others not. Results from the literature also suggest strong dependency of LCA results on system boundary, functional unit, data quality and allocation methods chosen.

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http://dx.doi.org/10.1016/j.rser.2012.04.016

1. Introduction

Climate change and energy security have, for many nations, become two of the greatest challenges. As a result the production of renewable energy has increased with the aim to reduce our current dependency on fossil fuels. Bioenergy, often with the benefit of little additional point of use infrastructure and non-immediate dependency on weather, offers a unique source of renewable energy. In particular, bioenergy from lignocellulosic biomass offers the potential to provide a significant source of clean, low carbon, and secure energy. Although often regarded as carbon neutral process there are environmental impacts associated with the system production, transportation, and growth of feedstock. As a result, sustainability assessment is now acknowledged to be an important element of development of bioenergy from lignocellulosic material.

One way to determine the impact of bioenergy is to use the environmental management tool, Life Cycle Assessment (LCA). LCA is a methodological tool used to quantitatively analyse the life cycle of a product or an activity within a generic framework provided by ISO 14040 and 14044 [1,2]. It examines the environmental burden of a product or process over its entire life, from production, through use and on to disposal or recycling. It consists of four methodological steps: goal and scope definition, inventory analysis, impact assessment and interpretation. Within the goal and scope the function of the system, its study boundary and environmental issues to be considered are identified. The system boundaries outline which processes and materials will be included and excluded from the system. The functional unit (FU) expresses the function of the system studied in a quantitative manner. The functional unit can therefore facilitate a direct comparison between different systems. The inventory stage is where data about the energy and materials used, and pollutants or wastes released into the environment as a consequence of a product or activity, are gathered. When a process produces more than one output an allocation procedure is used to allocate the environmental burdens between main products and co-products. After the inventory data are gathered an impact assessment is carried out to transform the long list of inventory results into the potential impact upon a limited number of environmental issues. These issues, or indicators, are classified into categories according to their potential long term damage such as climate change and ozone depletion. The indicator scores express the potential relative severity of the product or system examined on an environmental impact category.

By using LCA to examine the system of interest, quantifying the material and energy inputs and outputs to air water and soil, the potential impact on the environment can be determined. To date several studies [3–6]; have examined the environmental impact of bioethanol, with a particular focus on two main categories: greenhouse gas (GHG) emissions and fossil energy efficiency. These studies show, to a varying degree, reduction of fossil fuel use and of GHG emissions in comparison with the use of conventional energy such as gasoline.

Biomass is often considered to be a carbon neutral feedstock but a significant amount of GHG emissions are released during the life cycle, for example as part of the fertiliser production and use, during the transportation of the biomass, as well as in the conversion stages. Additionally, comprehensive sustainability assessment of biofuel is urgently needed to assess economic, social and environmental impacts of biofuel production and consumption [7]. Yan and Lin [8] revealed that the interactions among various sustainability issues make the assessment of biofuel development difficult and complicated. In addition, the complexity of the whole biofuel production chain can generate significantly different results due to differences in input data, methodologies applied, and local geographical conditions.

Lignocellulosic biomass can be converted to ethanol through feedstock handling, pretreatment, hydrolysis and fermentation, ethanol recovery and wastewater treatment [9]. Fig. 1 shows a typical process of the bioethanol conversion system. A typical LCA study of biofuel includes the feedstock growth at farming level, biofuel conversion process, and fuel use in transportation stage. Although LCA work [3] has shown environmental benefits associated with lignocellulosic ethanol, most studies have focused on assessing the farming systems with generic assumption of the ethanol conversion process. Very few have addressed the specific environmental issues related to the conversion process due to process uncertainties and non-availability of commercial scale plant [10]. Despite extensive research on laboratory and small scale within the scientific community, there is not yet a large scale commercial lignocelluloses-to-ethanol facility. Therefore technology uncertainty and potential commercial scale operation parameters also contribute to the gap [10].

The UK and EU is committed to producing 10% biofuel for transportation by 2020 [11]. Therefore the production of bioethanol is of great interest to energy suppliers and vehicle manufacturers. It is important that we meet our targets with the minimal impact on the environment; therefore a review of the current knowledge in this area is desirable. This paper reviews the existing literature of LCA studies for lignocellulosic ethanol processes with an aim to identify research gaps and therefore where future research should focus. The paper starts with a brief statement of study approach, and then provides an overview of the LCA studies reviewed in this paper, and follows a summary of

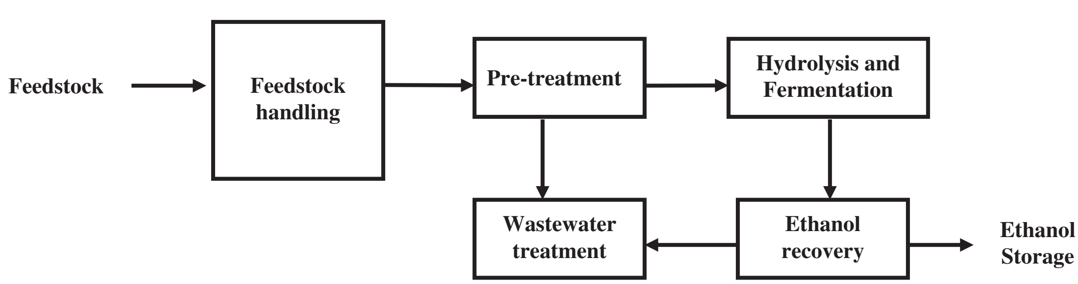

Fig. 1. Typical lignocellulosic ethanol conversion process.

key findings and research gaps with conclusions and recommendations for future studies.

2. Approach

The objective of the study was to review recent evaluations of bio-ethanol, made from various lignocellulosic feedstocks for use as a transportation fuel, compared to conventional fuels on a life cycle basis. This paper therefore consists of a literature search, a study of the methods and assumptions used, followed by an analysis of results and findings presented. Some results were normalised for the purpose of comparison; detailed information about how data were normalised is presented in the relevant sections.

An online search of scientific manuscripts was conducted to find studies that have been published in recent years (between 2005 and 2011). Previous literature including LCA of both 1st and 2nd generation ethanol has been reviewed by von Blottnitz and Curran [12]. The focus of this search was on the LCA studies of the conversion of lignocellulosic materials to ethanol processes.

Literature was searched using the tools Engineering Village, Google Scholar and Web of Knowledge. To maximise the number of literature captured, key words – Life cycle assessment, GHG, environmental impact, and ethanol were used. Papers were then selected for this review if lignocellulosic feedstock were examined. Lignocellulosic feedstock in this case means waste or agriculture residues, or dedicated plant biomass such as energy crop, switch grass. Algae biomass feedstock is not within the scope of this study. Feedstock is defined as raw materials to supply the ethanol conversion processes. In total, 53 scientific manuscripts containing LCA studies of lignocellulosic biomass and another 15 manuscripts with general overview or sustainability issues were reviewed for this paper.

3. Second generation ethanol conversion process

Conventional bioethanol, also known as "first generation", is produced from sugar, starch or oil biomass feedstock mainly cultivated from crops that can be used for food, but can also be grown as dedicated energy crops, such as sugarcane in Brazil. About 3.4 billion gallons of ethanol are generated annually from cane sugar in Brazil, but at currently controlled levels, prices are too high for sugar to be a viable feedstock in the UK [13]. The barriers of first generation biofuels (e.g. competition with food, high energy inputs, poor energy balances, low yields per hectare, and damage to ecosystem) can be partly overcome by the utilisation of second generation biofuels.

Second generation ethanol is made from lignocellulosic materials such as forestry and agricultural residues, significant portions of municipal solid waste (e.g. paper waste and yard waste), and woody and grassy crops grown to support fuel production. Lignocellulosic materials have numerous advantages, including sometimes being a co-product of food production (e.g. straw) and often being available in large quantities. The material can also be used to produce other high value products such as chemical compounds, electricity and heat [14]. Hence, second-generation bioethanol is expected to make a major impact on transportation fuel markets, without some of the negative impacts associated with first generation fuels.

However, lignocellulose is a more complex substrate than starch. It is composed of a mixture of carbohydrate polymers (cellulose and hemicelluloses) and lignin. The carbohydrate polymers are tightly bound to lignin mainly by hydrogen bonds but also by some covalent bonds. The biological process for converting the lignocelluloses to fuel ethanol requires the following: delignification to liberate cellulose and hemicelluloses from their complex with lignin, and depolymerisation of the carbohydrate polymers to produce free sugars to produce ethanol [15]. The delignification of lignocellulosic raw material is the rate-limiting and most difficult task to be solved. Another problem is that aqueous acid used to hydrolyse the cellulose in wood to glucose and other simple sugars destroys much of the sugars in the process. Extensive research has been carried out in this field over decades [16].

Nearly all of the ethanol fermentation technologies use mechanical pre-processing to remove recyclables and contaminants, shredding of the materials and drying to certain moisture content. Then the material is processed further in order to make use of cellulose and hemicellulose. Depending on the technology, this may include high temperature, acid treatment, and/or high pressure. Following the initial hydrolysis phase, the slurried materials are then fermented to produce alcohol, which is then purified through distillation and/or filtration to produce the desired fuel-grade quality ethanol. However, despite research attention on the technology development, the industrial scale up of this process appears to be still hindered by technological issues or by the lack of a biomass refinery approach [4]. This lack of industrial scale and subsequent technology uncertainties contributes to the issues of data uncertainty within LCAs.

4. LCA study of ethanol conversion process

Table 1 shows the 53 reviewed LCA review papers that examined the production of ethanol from lignocellulosic sources. All of the papers reviewed were published between 2005 and 2011. In addition, a number of studies [12,62–65,74] aiming at identify key issues are included in the discussion in the following sections. The LCA papers reviewed show different results due to variation in data, system boundary selection, and in methods used.

Bioethanol is often used as a blend with conventional gasoline, for example a blend named E10 is 10% ethanol mixed with 90% fossil based gasoline. Within this review paper a number of blends were examined. In addition, a number of differing environmental issues were included in the paper, from the more common energy consumption and GHG emissions, to other environmental issues such as acidification, eutrophication (over nitrification of waterways) and ecotoxicity and human health impacts.

Overall the studies reviewed show a range of GHG savings from 4% to 15% when shifting from conventional gasoline to E10, from 12% to 96% with E85, and from 46% to 90% with E100. The energy saving varies from 4% to 8% when moving from gasoline to E10, from 45% to 76% with E85, and 56% to nearly 100% with E100. Contrary results for the impact of acidification and eutrophication potential from lignocellulosic ethanol are also observed—some report less impact in comparison to conventional gasoline whilst others report up 800% and 1700% increase of acidification and eutrophication potential by ethanol production respectively. Studies also show the water consumption varies significantly depending on the types of biomass, requirement for irrigation, and regional irrigation practices; with different findings on whether agricultural practices or ethanol conversion are the main sources for water consumption. Mixed findings on emissions contributing to ecotoxicity and human health have also been reported with some being favourable with lignocellulosic ethanol while others not.

This paper analyses the literature between 2005 and 2011. Previous literature between 1996 and 2004 was reviewed by von Blottnitz and Curran [12] who carried out a review on the sustainability assessment of ethanol from both sugar crops and

Table 1
Summary of LCA studies of lignocellulosic ethanol.

Indicators	Biomass type	System boundaries	Functional unit	References
GHG & Energy	CS and WS	Well to wheel	The amount of agricultural residues treated per year	[17]
	Woodchips	Cradle to grave	Production of 4 m^3 of hardwood chips	[18]
	CS, switchgrass	Cradle to gate		[10]
	Cassava, sweet sorghum	Well to wheel	1 MJ fuel	[19]
		Fuel cycle	The amount of Fuel (MJ) to drive for 1 km	[20]
	MSW	Well-to-gate	1.2 gallons per year	[21]
	Poplar, CS, WS, Waste paper	Well to gate	1 litre of ethanol	[22]
	CS, FS, and switchgrass	Well to Wheel	g CO_2 eq of fuel	[23]
	MSW	Well to Tank	kg CO_2 eq per Litre of ethanol	[24]
	MSW	Well to gate Well to wheel	MT MSW; 1 km driven	[25]
	Crop residues	Well to wheel	kg CO_2 eq per km travelled	[26]
	Carob pod	Well to gate	1 kg ethanol produced	[27]
	Carob pod	Well to gate	1 kg ethanol produced	[73]
	Cassava, molasses	Well to wheel	1 MJ fuel	[74]
GHG emission	CS and corn	Well to gate	1 MJ energy	[28]
	MSW	Cradle to gate Cradle to grave	Total amount of waste treated MJ of fuel eq	[29]
	Wheat straw	Biomass supply	kg CO_2 eq/day	[30]
	Straw and CS	Well to gate	CO_2 emissions per litre of fuel grade ethanol	[31]
	Forest biomass	Well to Wheel	CO_2 eq per km travelled	[32]
	Poplar	Well to Tank Well to Wheel	1 MJ fuel 1 km driven	[33]
	Switchgrass	Well to Wheel	1 km driven	[34]
	Willow	Well to Wheel	1 tonnes ethanol	[35]
	Switchgrass	Well to Tank	1 MJ of ethanol	[36]
	CS, WS, Switchgrass	Well to Wheel	1 km travelled	[37]
Energy consumption	Lignocellulosic biomass	Cradle to gate	The amount of energy required to produce 1 litre of fuel	[38]
	Corn stover	Well to product	1 L ethanol	[39]
	WS, CS	Cradle to gate	Energy savings per tonne chemicals, per tonne harvested biomass feedstock and per cultivated land area	[40]
	Banana residues	Well to gate	1 L ethanol produced	[41]
	Corn stover	Well to gate	1 L ethanol produced	[42]
Water use	Switchgrass	Well-to-wheel	Gallons of water consumed per vehicle mile travelled	[43]
	Bioenergy crop	Well to gate	L water consumed per ha, per MJ fuel	[44]
	Poplar, CS, WS Waste paper	Well to gate	1 L ethanol	[22]
	Miscanthus and CS	Well to wheel	L water per km travelled	[45]
	Switchgrass	Well-to-wheel	1 L ethanol	[46]
CML indicators	Poplar biomass	Cradle to wheel	1 km distance driven by a middle size Flexi fuel vehicle	[47]
	Lignocellulosic feedstocks	Well-to-wheel	1 km distance driven by a FFV	[48]
	Alfalfa stem	Well-to-wheel	1 km distance driven by a FFV	[49]
	Brassica carinata	Well-to-wheel	1 kg of pure ethanol, 1 km driven by an ethanol-based fuelled vehicle	[50]
	Cassava, Molasses	Cradle to gate	1000 L of ethanol	[51]
	Flax shives	Well to gate Well-to-wheel	1 kg of pure ethanol, 1 km driven by an ethanol-based fuelled vehicle	[6]
	Corn stover	Well-to-wheel	1 km driven by a midsize car	[5]
	Black locust	Well-to-wheel	Kg CO_2 eq per 545 km distance travelled by a full tank of gasoline	[52]
	Switchgrass	Well-to-wheel	The amount of biomass treated per year	[53]
Impact 2002	Sugarcane bagasse	Cradle to gate	The sugarcane bagasse harvested from a hectare of land per year	[54]
EcoIndicator 99	Sugarcane bagasse	Cradle to gate		[55]
	Straw	Cradle to gate	1000 kg Straw	[56]
Other sustainability criteria	Willow	Biomass production	per oven dry tonne of biomass produced	[57]
	virgin timber resources, recycled newsprint	Cradle to gate	1 L ethanol produced	[4]
	MSW	Well to Tank	g H+ per Litre of ethanol produced	[24]
	Bio-based feedstocks	Cradle to grave	The risks related to the use of 1 ton of maize, expressed in YOLL	[58]
	Karlstad Salix	Cradle to gate	One site (5000 m^2)	[59]
	Switchgrass	Cradle to grave	1 km driven by a middle size car	[60]
	Forrest wood	Cradle to grave	1 km by E85	[61]

Note: CS—corn stover, WS—wheat straw, FS—forest residues, MSW—municipal solid waste.

lignocellulosic material. Their review discussed results in three categories of special interest to the question of environmental sustainability: (1) reducing dependence on fossil fuels through energy balance assessments; (2) reducing emissions of greenhouse gases (GHGs); and (3) reducing health and environmental impacts throughout the life cycle. The main findings include that bio-ethanol results in reductions in resource use and global warming; however, impacts on acidification, human toxicity and ecological toxicity, occurring mainly during the growing and processing of biomass, were more often unfavourable than favourable. Whitaker et al. [79] also reviewed 44 LCA studies of first and second generation bioethanol; their study is limited to GHG and energy balance at well to tank level. In comparison to the metrics included in von Blottnitz and Curran [12] and Whitaker et al. [79], this review covers GHG and energy balance at well to wheel level for ethanol fuel produced from lignocellulosic material, water issues and in-depth analysis of acidification and eutrophication, system boundary and functional units, data sources, and allocation of by and co-product credits.

The conflicting results of LCA studies are often due to the use of different functional units, system boundaries and data. These differences often lead to difficulties in comparing studies. Published LCAs often focus on different system boundaries, such as well to tank or well to wheel, and different functional units such as 1 kg of ethanol, distance travelled with ethanol blend fuel, as well as using different off the shelf life cycle impact assessment methods, for example, CML [66] and EcoIndicator [67].

In terms of ethanol conversion process, it is also not clear in some studies what has been taken into account. The NREL simulated process based on corn-stover has been adopted by a number of LCA studies [48–50]; whilst others [27] have not specified the processes. It is also not always clear if chemicals, enzymes and infrastructure have been taken into account. Most papers that assess the conversion of specific feedstocks to bioethanol have not gone beyond energy and carbon assessments. However, the overall environmental sustainability of bioethanol cannot be assessed without investigating other environmental and socio-economic impacts [29].

The results of this review are discussed in four categories: (1) LCA methods and impact indicators; (2) System boundary and functional unit; (3) data source; and (4) allocation methods. Each interest area is discussed in more detail in the following sections with the aim to identify gaps and challenges in LCA studies of lignocellulosic ethanol production.

4.1. System boundaries and functional unit

4.1.1. System boundary

Defining system boundaries is critical to appropriately conducting an LCA study. Although the system boundaries of biomass ethanol can vary from study to study depending on the inclusion or exclusion of some specific processes, a comprehensive LCAs would consider [68]: (i) the production of inputs used in growing biomass (e.g. corn, wheat), including seed, fertilizer, herbicide, pesticide and energy; (ii) the application and utilisation of inputs and the harvest of biomass feedstock; (iii) the transportation of biomass to biorefineries; (iv) the conversion of biomass feedstock to ethanol at biorefineries; and (v) the eventual burning of ethanol as transportation fuel.

The system boundary of well to tank (also called as well to gate, cradle to gate) omits the fifth stage. For example, for well to tank, studies [31,38] often use kg or L ethanol as functional unit. For well to wheel, travelled distance or amount of energy are used [19,32,33]. Both systems have advantages and disadvantages. Depending on the study purpose, some studies [20,57] have only considered biomass supply chain or fuel cycle in the end use.

Not all studies take into account the chemicals, enzymes, nutrients, and the infrastructure (such as equipment) required. As pointed out by MacLean and Spatari [36], there is a gap in lignocellulosic ethanol LCA studies; most published studies have not accounted for the impacts associated with the production and use of pre-treatment chemicals, enzymes and nutrients used in the conversion process. They further report that a third of GHG emissions over the life cycle are contributed to enzymes and chemical. Kemppainen and Shonnard [4] conducted a comparative life-cycle assessment for biomass to ethanol production from different regional feedstocks. Their study investigates the effect of process improvement on the life cycle impact of lignocellulosic ethanol at cradle to gate level and highlights that the environmental impacts from the pre-manufacturing process and the manufacturing process of ethanol must be quantified if the complete benefits are to be determined [4]. One study [27] reported up to 70% saving of GHG emissions, but the study did not take into account machinery used in the biomass growth stage or infrastructure use during the ethanol conversion processes.

The variation in system boundaries makes it difficult to easily compare results in the literature. More common boundaries and transparent information, including data for sub steps (for example the data per litre followed by the data per distance travelled) would enable readers to clearly see the difference between studies.

4.1.2. Functional unit

The functional unit is the central core of any LCA and is the reference unit that forms the basis for comparisons between different systems. Results from different types of functional units considered (e.g. ethanol production, or travelling distance based) are slightly different from each other in most of the environmental impact categories. Gonzalez-Garcia et al. [6] illustrates that the results of an LCA are strongly dependant on the chosen functional unit by using two types of functional unit in the same study with same system boundary and data input. They compare 1 kg of pure ethanol and 1 km driven by an ethanol-based fuelled vehicle: comparing both ethanol blend fuel E10 and E85, for ethanol production oriented function unit (1 kg ethanol) E85 offers environmental advantages such as acidification and eutrophication when compared to E10; while for travel distance oriented functional unit (1 km), the results are contrary.

Table 1 includes an overview of the functional units used in the reviewed LCA studies. In most of the articles investigated the functional unit was based on mass such as kg of ethanol [6,27], on volume e.g. L of ethanol [22,41,42] or on driven distance e.g. km [25,32,34] and according to their results, the selection of the functional unit considerably affects the final conclusions. Other functional units are not directly linked to fuel or energy produced, such as volume of hardwood chips [18], the amount of waste treated [17,29], and per oven dry tonne (ODT) of biomass produced [57] can make results difficult to compare as fuel production largely depends on the type of biomass, feedstock composition as well as conversion rate. Cherubini et al. [26] suggests that LCA results of transportation biofuel production should be expressed per km basis, in order to take into account engine mechanical efficiencies, type of fuel and emissions from combustion (which are relevant for fossil reference systems based on conventional fossil fuels) and states that results reported per 1 MJ of fuel can be misleading. However, it can also be argued that a volume or energy related functional unit is better as emissions and efficiencies vary significantly between different vehicles.

4.2. LCA and environmental impact indicators

Most of the impact assessment methods presented in the reviewed papers focus on energy and carbon accounting; only a few studies attempt to be more inclusive in addressing sustainability of a wider range of environmental issues. von Blottniz and Curran [12] concluded that that resource demand and GHG emissions are generally lower for bioethanol production. However, they suggest that more attention should be placed on impacts on human health and ecological risk due to biomass bioethanol systems. Biobased biofuel systems have possible ecological drawbacks: agricultural production of biomass is relatively land, and sometimes water, intensive, and there is a risk of pollutants entering water sources from fertilisers and pesticides that are applied to the land to enhance plant growth. These are not often considered within the studies reviewed (10 out of 53 studies reviewed considered human health and ecotoxicity related issues). Among all the studies reviewed, more than half of the studied manuscripts focus either a single issue of GHG emissions, or energy consumption, or a combination of both. While GHG emissions and energy savings are the centre of attention in many LCA studies (e.g. [29,34,69]), very few studies have considered potential impacts on acidification, eutrophication and ozone creation potential [5]. However, some of these wider environmental burdens are site specific, thus limiting generalisation of the results [26].

4.2.1. GHG emissions

All but two of the studies reviewed conclude that there is a reduction of GHG emission when using ethanol from lignocellulosic feedstock in comparison to fossil fuel reference system. Fig. 2 shows the range of the GHG emissions from different biomass sources published in the literature. Only those studies considered cradle to grave or well to wheel life cycle are compared. Some data presented as function unit of kg ethanol and MJ ethanol are harmonised to per km distance travelled with the assumption that a distance of 1 km can be travelled with 2.5 MJ of pure ethanol. Fig. 2 shows that when shifting from conventional gasoline to E10, the GHG emissions range from −1.10 kg CO_2 eq/km travelled to 0.28 kg CO_2 eq/km, −1.15 Kg CO_2 eq/km to 0.79 kg CO_2 eq/km for E85, and −1.25 KgCO_2 eq/km to 0.84 kg CO_2 eq/km, in comparison to 0.26 Kg CO_2 eq/km from conventional gasoline.

Two studies report higher GHG emissions from ethanol blend fuels with switchgrass and corn stover than conventional gasoline when economic value allocation is applied. However, in the same studies, when other type of allocations such as mass allocation and system expansion are applied, GHG emissions are lower than conventional gasoline in all types of ethanol fuel blends. The influence of the allocation method in the results is discussed in the Section 4.4. Three studies [5,49,50] report negative GHG emission: one [5] found negative GHG values when using system expansion allocation—this is because the study takes into account C sequestration from both food (corn) and co-products (corn stover) whilst within the studied system only CO_2 release from coproduct (corn stover) to ethanol are counted. The other two [49,50] report negative GHG values when higher ethanol blends (i.e. E85 or E100) are used—this is because the GHG emission from the whole life cycle is smaller than the carbon sequestration during biomass growth resulting negative GHG values.

Different GHG emission sources have also been reported. Wang et al. [23] found that for switchgrass-based cellulosic ethanol, GHG emissions from nitrogen fertilizer in farms are the largest source, while Gonzalez-Garcia et al. [49] argues that the GHG contributions are from the ethanol production and then followed by fuel blend use. Kemppainen and Shonnard [4] also suggest that the climate-active carbon dioxide is primarily due to the pre-manufacturing life-cycle stages of chemicals used in the process and the use of utilities in the ethanol production process for newsprint. Maclean and Spatari [36] investigated the sensitivity of fossil energy and GHG impacts from various process conditions, conversion technologies, and enzyme loadings and found a much greater contribution from enzyme production than another study [24], with 33%–35% contributions to the well to wheel life cycle.

Gonzalez-Garcia et al. [52] carried out a detailed analysis of the GHG emission and highlight the relevant role of two global warming gases—CO_2 and CH_4. The contributions of CO_2 represent the 98%, 99% and 86% of total GHG emissions for conventional gasoline, E10 and E85, respectively; CH_4 contribution adds up to 13% in E85; and the contribution from N_2O emission (mainly from agricultural subsystem) is insignificant. The findings are in contrast to other related studies [49,60] where N_2O emissions are

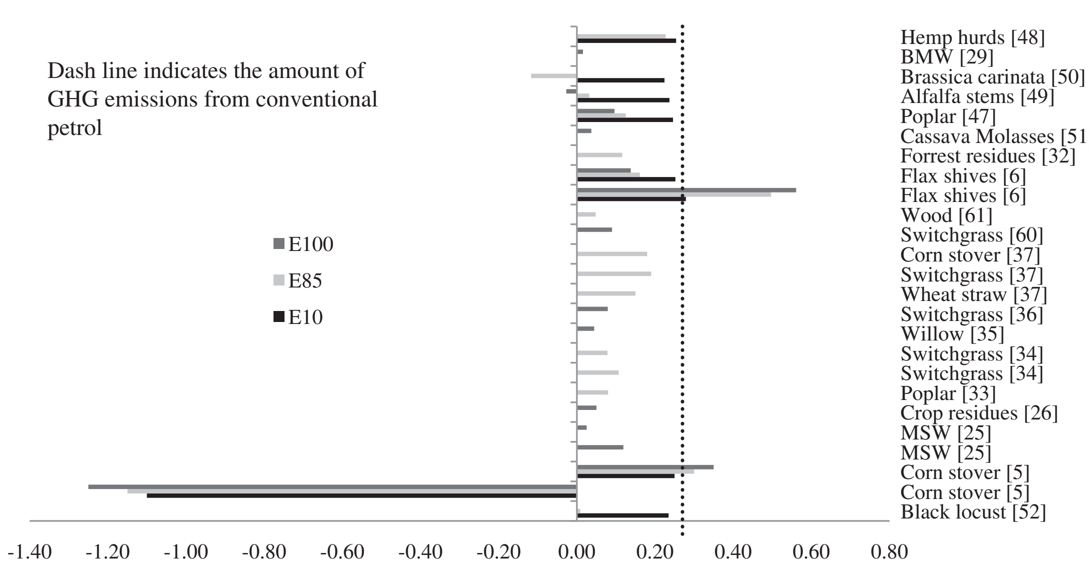

Fig. 2. Comparison of GHG emissions per km travelled with different ethanol blend fuel (unit: kg CO_2 eq/km). *Note*: MSW—municipal solid waste, BMW—biodegradable municipal waste.

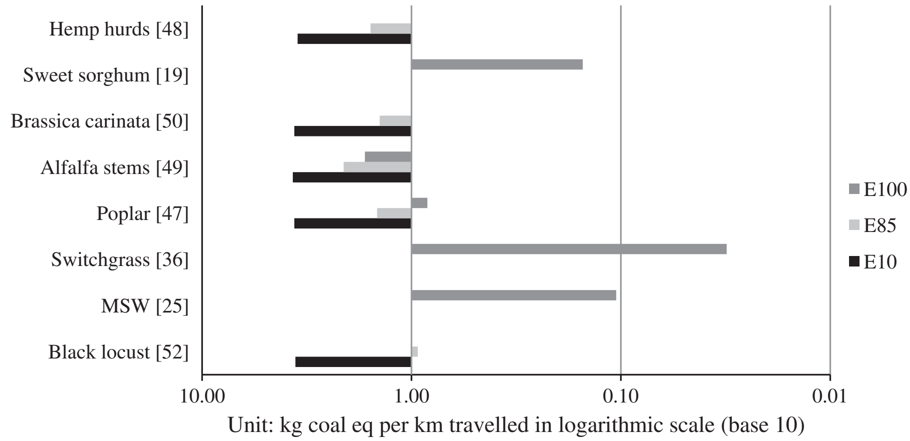

Fig. 3. Fissile fuel consumption per km travelled with different ethanol blend fuel (unit kg coal eq/km). *Note*: MSW—municipal solid waste.

found to reach 15% of total GHG emission. Zah et al. [70] also shows that the most relevant factors for the greenhouse gas emissions at farm level are agricultural N_2O emissions and CO_2 from land transformation.

4.2.2. Energy consumption

Similar to GHG emission, energy efficiency has been one of the key issues studied by the scientific community. Although the energy savings calculated are different in the reported studies, energy savings were reported in all cases. Fig. 3 shows the range of fossil energy used that have been reported for ethanol blend fuels E10, E85 and E100. Only data from those studies considering well to wheel or cradle to grave life cycle are presented in Fig. 3. The majority of the studies reviewed at well to wheel level report energy consumption as kg coal eq per km travelled. Some studies [25,36] report a unit of MJ fossil energy per MJ ethanol; as a result those data in MJ fossil energy per MJ ethanol are converted to the unit of kg coal eq per km travelled for the purpose of comparison with other studies. It was assumed that 1 kg coal is equivalent to 24 MJ energy and 2.5 MJ of ethanol is required to run a medium sized car for 1 km. Fig. 3 shows that the amount of fossil energy consumed to produce E10 ranges from 3.5 kg coal eq/km to 3.69 kg coal eq/km, corresponding a saving of up to 8.6% in comparison with conventional petrol. Similarly, for E85, a saving of up to 76% can be achieved and nearly 100% savings for E100 for the best case reported.

Studies on well to gate life cycle have also reported fossil energy savings. For example, Kaufman et al. [28] reports up to 80% energy savings in biorefinery ethanol production in comparison to a reference fossil fuel system. Kemppainen and Shonnard [4] state that the amount of life-cycle fossil energy required to produce ethanol is 14% of the energy content of the product, making the overall efficiency 86%. Schmitt et al. [24] and Maclean and Spatari [36] finds that enzymes and chemicals input for the ethanol conversion process make up 30% of both total fossil fuel input.

However, a study carried out by Ojeda et al. [55] showed a net energy ratio lower than 1 by conducting energy analysis of four second generation ethanol conversion technologies including acid dilute hydrolysis, liquid hot water, acid catalysed with steam explosion and organosolv solution. The study suggested that heat integration methodologies are necessary to improve energy efficiency in these processes. In contrast, Luo et al. [5] stated that when all the co-products are taken into account in the corn stover-based ethanol, the net energy value becomes much higher than that cited in the literature covering the corn cases, which shows ethanol production from cellulosic feedstock is more energy efficient than corn-based ethanol. Nevertheless, the articles reviewed here suggest that when comparing to first generation ethanol such as those from corn, lignocellulosic ethanol offers much better reductions in fossil energy consumptions.

4.2.3. Acidification and eutrophication

Acidification and eutrophication are reported in some of the studies, predominantly those that use the "off the shelf" impact assessment methods such as CML and Impact 2002. Figs. 4 and 5 summarises the findings of acidification and eutrophication potential reported in the literature. Among the seven studies reporting acidification, two suggest lower impact of acidification potential in comparison with conventional gasoline; while five show opposite findings in all case of ethanol fuel blends. Gonzalez-Garcia et al. [52] reported that it was possible to reduce the acidifying emissions by increasing the ratio of ethanol in the blend using black locust as biomass. Similar findings are also reported by Luo et al. [5] using corn-stover as feedstock. These two findings are completely different when compared to other related studies [6,47,48,60].

One study [47] on poplar biomass reports nearly 800% increase of acidification potential when comparing E100 to conventional gasoline. Similar findings within the same study [47] are reported for eutrophication potential with a 1700% increase when compared E100 with conventional gasoline. It appears that the increase of ethanol portion in the fuel blends results in an increase of emissions for both acidification and eutrophication.

Different reasons for the increase of these indicators have been reported: Cherubini and Ulgiati [17] suggest the increase is mainly due to N fertilisation, which causes leaching of nitrates to groundwater; Schmitt et al. [24] suggests that nearly 90% of acidification was from sulphuric acid production alone.

4.2.4. Water

In recent years a number of studies have placed attention on water issues for biofuel production. Fig. 6 shows the water consumption in relations to lignocellulosic bioethanol production at life cycle level reported by the literature reviewed. Data was expressed as L water consumed per km distance travelled by using E100. Fig. 6 shows a clear variation of water consumption depending on the types of feedstock ranging from one L to nearly 14 L per km travelled with E100. This is further confirmed by Fingerman et al. [44] suggesting the water consumption varies by up to 60% among different feedstocks. Within the same study, it was also found that the amount of water required producing ethanol range from under 500 to over 3500 L water per L fuel. The

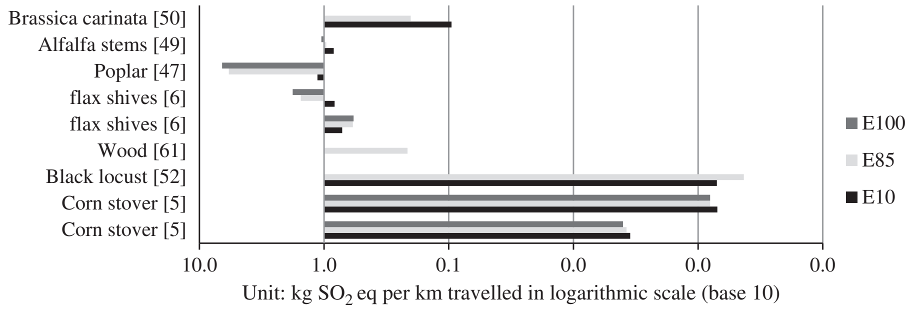

Fig. 4. Acidification potential from different ethanol blend fuel (unit: kg SO_2 eq/km).

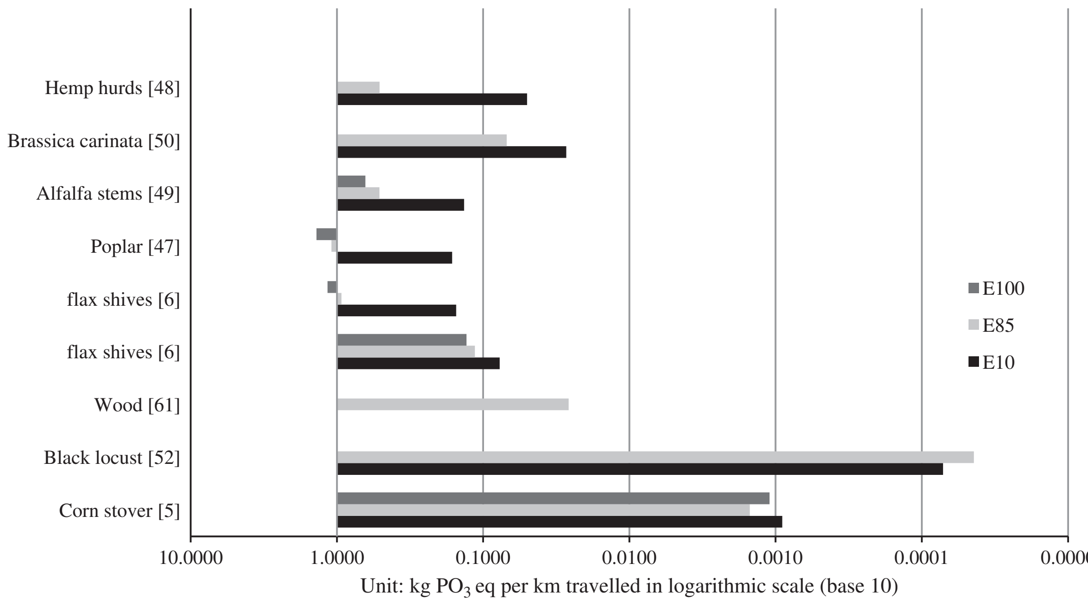

Fig. 5. Eutrophication potential from different ethanol blend fuel (unit: kg PO_3 eq/km).

resulting burden from biofuels production is highly dependent on whether the crop requires irrigation and the regional irrigation practices [45,71].

Differing options for the source of water consumption during the ethanol life cycle have been observed in the literature. Mu et al. [22] concluded that more than 90% of water consumption comes from ethanol plant; however, Fingerman et al. [44] proposes that life cycle water consumption for ethanol production is due to a cultivation phase that consumes over 99% of life cycle water use for agricultural biofuels. Wu et al. [46] reported both reasons—crop irrigation and ethanol processing for the large water consumption during the life cycle of ethanol production; and it was suggested that with the technology improvement, the water consumption can be reduced to 5.9 L/km travelled with E100 from 9.8 L with current technology.

Scown et al. [45] outlines out that biofuels may place a larger burden on groundwater than electricity on gasoline production, whereas the latter depends more heavily on surface water. Water use also has an impact on the increase in GHG emissions that result from energy used for pumping and treating water for irrigation, cooling, mining/extraction, and industrial use [45]. This climate impact needs to be considered before placing biorefineries in areas requiring desalination, wastewater recycling, or water importation. The potential water related GHG emissions is 180 g CO_2 eq per km travelled for corn grain and stover based ethanol [45,64]. In general, data including water use and water resource required for analysing the additional climate impact associate with water supply is lacking. According to Scown et al. [45] mining/extraction and industrial water requirement information is particularly scarce and water resource information in particular with respect to groundwater is lacking. Sheehan [64] outlines that another problem for biofuels and water is that the impact of increased biofuels production on water quality, mainly with regard to nutrient leaching and eutrophication from farming operations, is poorly understood. Life cycle studies to date do not adequately account for these effects.

4.2.5. Ecotoxicity and human health

A number of indicators contributing to human health and ecotoxicity have also been reported in the literature. Table 2 shows ten studies which aim to cover a wider range of indicators beyond GHG and energy issues. Because not all indicators are used by every study, NA in Table 2 indicates 'not applicable' for such indicators in the appropriate studies; "+" means an increase of impact from lignocellulosic ethanol has been observed in comparison with fossil fuel reference; "−" suggests a decrease of impact from ethanol production compared with reference system; and "±" indicates both increase and decrease are presented depending on allocation methods. In general, all the studies reported suggest an increase in the impact on

photochemical oxidants formation by lignocellulosic ethanol production. Similar trends have also been reported for ecotoxicity except in one study. Contrary findings have been reported for human toxicity with half suggesting a decrease of impact by lignocellulosic ethanol. All but one [6] of the studies indicates a decrease in ozone depletion. A decrease in abiotic depletion is also found by all studies reporting this issue. The study by Uihlein and Schebek [56] also reports decrease for ionising radiation, and respiratory effects but increase in carcinogenic.

The observation shown in Table 2 is different from the finding by Roes and Patel [58] who indicate that that the conventional risks of all bio-based products are lower than those of the petrochemical products, and that the chemical industry sector contribute more to the final risks of petrochemical products than bio-based products. They also indicate that the agriculture sector was only of relevance to biobased products. Luo et al. [63] suggest the main contributors of human and eco-toxicity are the production of chemicals and machinery used in agriculture. Using agricultural residues such as corn stover has less impact in human and ecotoxicity than using energy crops such as switchgrass; the reason for this is again the partitioning factors based on economic allocation in the agriculture of corn stover, flax shives, and hemp hurds. Large amounts of emissions in these three cases are allocated on the main crops: corn, flax, and hemp.

Sheeham [64] states that ecosystem health and biodiversity is another aspect of biofuels that remains largely unstudied. However, Ojeda et al. [55] compared different technology scenarios of ethanol conversion process and found that impacts over ecosystem quality were observed in all cases. Kemppainen and Shonnard [4] argue that the challenge of ethanol from biomass such as woodchips is to determine the ecological impacts of large scale timber harvesting, a topic not yet studied. Suer and Andersson-Skold [59] suggest that further development of impact assessment methods is necessary, especially with regard to biodiversity and land surface as a limited resource.

This review shows that a number of environmental issues need to be studied further. The authors believe that when considering alternatives to fossil fuels we must better understand not only the cost and carbon impact but also potential impacts on human health, natural resources, and other environmental impacts such as air pollution and eutrophication. Some studies [29,77] also state that the overall environmental sustainability of bioethanol cannot be assessed without considering other environmental, economic impacts and production scale effects.

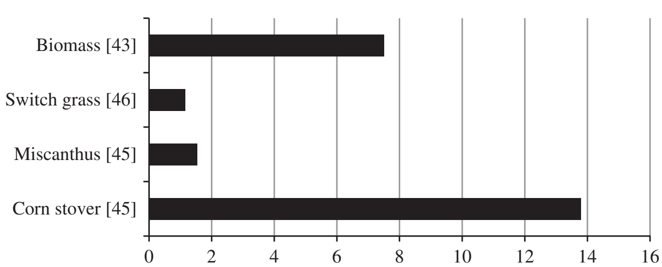

Fig. 6. Water consumption for lignocellulosic ethanol production (unit: L water/km).

4.3. Data sources

Reliability of the results from LCA studies strongly depends on the extent to which data quality requirements are met. For LCA studies of lignocellulosic ethanol conversion process, data such as material flow, energy flow, and infrastructure of industrial scale ethanol conversion plant are needed. However, due to the lack of commercially available data; studies taking into account the manufacturing processes often rely on simulation data [9]. In the case of simulation-based LCA studies, assumptions have to be made according to the simulating model chosen. MacLean and Spatari [36] state that there is a gap in many lignocellulosic ethanol LCA studies: most published studies have not accounted for the impacts associate with the production and use of pretreatment chemicals, enzymes and nutrients used in the conversion process. A common problem in evaluating competitive technologies is the difference in degree of development, and hence reliability of data, for the technologies; most research in enzymatic hydrolysis of straw/stover followed by fermentation to ethanol is under laboratory investigate with a few in pilot plant hence detailed design data is not available in the literature [31]. Cherubini and Stromman [75] also highlighted the problem with data scarcity of advanced conversion technologies that hinder comprehensive LCA. The few existing studies are mainly approximations based on mass/energy balances.

Johnson et al. [78] show that the quality of data has a large impact on the variability of LCA results and it was suggested that data availability is the primary constraint dictating how uncertainty should be presented when modelling emissions from bioenergy systems. Cherubini [76] suggests a sensitivity or uncertainty analysis to be carried out as part of a LCA study to estimate the effects of variations in key parameters to the outcome of the study and identify the parameters with the largest influence on the final results and check the accuracy of those data.

Bright and Stromman [61] point out that for environmental impacts such as acidification and eutrophication, the fate and transport of airborne pollutants contributing to these categories may vary by region, because the buffering conditions in specific regions may be different. Hence region specific data for the non global categories will provide a more complete picture of the environmental implications of ethanol production and its use.

Table 2
Human and ecotoxicity indicators.

Indicators	Black locust [52]	Straw [56]	Corn Stover [5]	Switchgrass [60]	Wood [61]	Flax shives [6]	Poplar [47]	Alfalfa stems [49]	Brassica carinata [50]	Hemp hurds [48]
Eoctoxicity	NA	–	+	+	NA	+	+	NA	NA	NA
Photochemical oxidation	+	NA	+	+	NA	+	+	+	+	+
Ozone depletion	NA	–	–	–	NA	+	–	NA	NA	NA
ionising radiation	NA	–	NA	NA	NA	NA	NA	NA	NA	NA
carcinogenic	NA	+	NA	NA	NA	NA	NA	NA	NA	NA
respiratory effects	NA	–	NA	NA	NA	NA	NA	NA	NA	NA
Mineral extraction	NA	–	NA	NA	NA	NA	NA	NA	NA	NA
Abiotic depletion	NA	NA	–	–	NA	–	–	NA	NA	NA
Human toxicity	NA	NA	+	+	–	±	+	NA	NA	NA

Note: NA: not applicable; +: increase of impact from ethanol production compared to gasoline; –: decrease of impact from ethanol production compared to gasolin; ±: both increase and decrease are presented depending on allocation methods.

Table 3
Comparison of different allocation methods [62].

Allocation methods	Mass-based method	Energy content based method	Economic value method	System boundary expansion method
Principles	Energy use and emission burdens are allocated among all products according to their mass	Energy use and emission burdens are allocated among all products according to their energy output shares	Based on economic revenue shares of individual products	Energy use and emission burdens of producing the substituted products are estimated; the estimated energy use and emissions are credits that are subtracted from the total energy use and emissions burdens of the biofuel production cycle
Application	Widely used in LCAs of consumer product and in some generic LCA models	Applicable where most of the products are used for their energy content purposes	Used by economist; it normalises all products to a common basis – their economic values, regardless of the purpose of their use	Applicable in dealing with multiple products. Tends to represent the actual effects of generating multiple products from a pathway
Limitation	Not applicable to product without mass such as electricity as byproduct from biorefineries	Not suitable when products have distinctly different uses. For example, starched based ethanol plants produced ethanol and animal feed; the application reflects neither the use of individual products nor the energy use and emissions of producing individual products.	Subject to great fluctuation of product prices; the actual energy use and emissions that accrue during a fuel pathway may not be related to the economic values of individual products.	The methods can be time intensive and resource consuming. When non-fuel products are a large share of the total output, the method generates distorted fuel-based results.

In addition, there is a gap in enzyme LCA data [10] as data for enzyme manufacture are not available in life cycle databases or published literature. Enzyme manufacture can vary in its energy input and emission outputs depending on the enzyme family and the energy mix in the location of manufacture [65]. Hence, there is a need to establish a database for enzyme manufacture.

Kemppainen and Shonnard [4] carry out a comparative life cycle environmental impact assessment using a combination of process simulation, an impact assessment software tool for the manufacturing process life cycle stage, plus a life cycle inventory database coupled with an impact assessment software tool for the pre-manufacturing process impacts of fuels, process chemicals, transportation, and some preparatory steps (wood chipping etc). The main advantage of this approach is that process alternatives are relatively easily to simulate and evaluate for environmental impacts, facilitating the comparison of process improvement and pollution prevention modifications.

LCA models coupled with mathematic models, and process simulation models could be the key in predicting the environmental impacts of the lignocelluloses-to-ethanol conversion process. Results would provide information and direction for improving the environmental performance in the early stage of planning. Tools such as data mining, sensitivity analysis and uncertainty analysis also offer to identify and potentially correct data integrity issues.

4.4. Co-products allocation methods

Co-product allocation is one of the most controversial issues in the development of LCA methodology, because the results of an LCA study can be significantly influenced by the choices of allocation methods. Although more and more attention has been paid to allocation methodology in the recent years, specific guidance is still lacking. Table 3 summarises three common approaches of allocating environmental burdens with their advantages and disadvantages. A few studies have attempted to address the allocation issue in LCA to deal with biofuel and its by-products (e.g., [5,6,62,72,80]). Depending on the method selected, the results can vary widely.

Within the bioethanol conversion system, allocation can occur in different ways, including: (1) allocation within main crops and residues as lignocelluloses; and (2) allocation within the biofuel pathways. In biofuel production, products besides biofuel such as chemicals are often generated. These products, referred as co-products or sometimes by-products, generally have significant commercial value and are part of the viability of biofuels themselves. Hence allocation is often necessary to take into account these products.

Gnansounou et al. [69] stated that the net GHG emissions of ethanol production may vary with allocation method adopted (mass, energy or carbon content or economics), with carbon content being the most favourable and economy being the least favourable. Although the ISO preference is for system expansion, Kaufman et al. [28], argued that for measuring carbon intensity, simply allocating emissions based on physical or economic partitioning may be preferable, particularly if corroboration with historic emission inventories is important. The system expansion approach may not be consistent with the long-term goals of these regulations.

Gonzalez-Garcia et al. [6] argue that allocation methods are essential for outcomes and decision-making. Using ethanol as transportation fuel could present better environmental performance than conventional gasoline in terms of global warming and fossil fuel consumption according to mass allocation. However, environmental credits could be achieved in terms of acidification, fossil fuel consumption and human toxicity according to economic allocation. Luo et al. [5] and Hoefnagels et al. [80] also reported contrary results are observed with different allocation methods.

It is probable that future lignocellulosic ethanol plants will be equipped with cogeneration units that produce both steam and electricity [62]. Thus, the process-purposed-based approach and mass-based approach will not be applicable [62]. Hybrid allocation methods have been applied in some studies for the soybeans-to-renewable diesel pathway [62].This combined the system boundary expansion to estimate energy use and emission credits for soy meal from soybean crushing units and the energy content based method to allocate energy products from the renewable diesel plants. This hybrid approach could also be applied in bioethanol production. In all cases, allocation procedure is an important issue in lignocellulosic ethanol conversion to be further studied.

5. Research gaps and challenges

This review shows that the end results of an LCA are dependent on several factors e.g. the systems boundaries, functional units and allocation procedure. It was also shown that the definition of the functional unit is crucial; the same studies using different functional units can lead to different results.

Moreover, the developing conversion technologies may differ in their feedstock requirements as well as process energy and

chemical inputs. As a result, they exhibit a range of life cycle energy and environmental performance: an aspect that needs to be further investigated. A list of research gaps has therefore been identified:

1. Studies should have some common boundaries or have transparent information regarding system boundaries so that results from different studies can be compared. Functional units should be carefully selected and perhaps multiple FUs could be shown within studies to aid future comparisons.
2. Studies should be broadened to cover a wider range of environmental issues. Energy efficiency and GHG emission are dominant in the published life cycle assessment of lignocellulosic ethanol production. Other issues such as water, ecotoxicity and human health are important issues to be considered. Future studies should provide clear information with regards to the contrary results presented in the literature such as acidification and eutrophication.
3. Additional GHG emissions from water supply, chemical and enzyme inputs during ethanol production stages should be taken into account to provide comprehensive information with respects to the climate change potential.
4. Data gaps for the life cycle assessment of lignocellulosic ethanol should be addressed. There is a need to establish a database for enzyme and chemical manufacturing. LCA studies should also account for the impacts of associated production and use of chemicals, enzyme and nutrients. Lack of available data from commercial second generation ethanol plants and the uncertainties in technology performance have made the LCA study of lignocellulosic ethanol conversion process particularly difficult and challenging.
5. By-product credit and allocation is an issue to be addressed. Biorefinery systems are characterised by multiple high value products; both bioenergy carriers and other materials. It will be necessary to address how the environmental burdens and benefits can be addressed between co-products. Studies are lacking in this area; future studies are encouraged to investigate this issue.

6. Conclusions

Concerns about sustainability and security of fossil energy, along with advances in biofuel technology simulated the interested in ethanol production from lignocellulosic material. Several studies carrying out life cycle assessment of lignocellulosic bioethanol production have been conducted in the recent years and these have been reviewed in this paper. It is agreed that second generation ethanol provides savings of both GHG emissions and fossil fuel use. However, a wider range of environmental issues should be considered to provide an overall view of the sustainability of lignocellulosic ethanol conversion.

The life cycle assessment of lignocelluloses based ethanol published covers both agricultural and waste residues and plant biomass. These studies are summarised in Table 1 according to their studied environmental indicators. The environmental advantages of biomass based ethanol, regarding gasoline substitution and GHG emissions mitigation, have been observed in all of them. However, environmental disadvantages in terms of acidification and eutrophication have also been highlighted; these impacts are mainly occurred during the harvesting and processing of the biomass. The findings of the environmental indicators were in line with those of the review study by von Blottnitz and Curran [12].

All but two of the studies reviewed conclude that there is a reduction of GHG emission and fossil energy consumption when using ethanol from lignocellulosic material in comparison to a fossil fuel reference system. However, different studies have reported different sources contributing to GHG emission through the whole life cycle chain: some report that the majority of GHG emissions come from the biomass cultivation stage; others argue that significant GHG emissions are from ethanol conversion process. Overall the studies reviewed show a range of GHG savings from 4% to 15% when shifting from conventional gasoline to E10, from 12% to 96% with E85, and from 46% to 90% with E100. They show energy savings from 4% to 8% when moving from gasoline to E10, and from 45% to 76% with E85, and 56% to nearly 100% with E100. Contrary results for the impact of acidification and eutrophication potential from lignocellulosic ethanol are also observed. Some report less impact in comparison to conventional gasoline whiles others report up 800% and 1700% increase of acidification and eutrophication potential by ethanol production respectively. Studies also show that water consumption varies significantly depending on the types of biomass, requirement for irrigation, and regional irrigation practices, with different findings on whether agricultural practices or ethanol conversion being the main sources for water consumption. Contrary findings on emissions contributing to ecotoxicity and human health have also been report with some being favourable with lignocellulosic ethanol while others not.

This review also finds a strong dependency of LCA results on system boundary, functional unit, data quality and allocation methods chosen. The lack of available data from commercial second generation ethanol plant and the uncertainties in technology performance have made the LCA study of the lignocellulosic ethanol conversion process particularly difficult and challenging. There are still many ongoing issues surrounding both the methodologies of LCA studies and data available. Without an transparent LCA, inconsistent system boundaries or input assumptions would most likely result in incomparability of different studies. For an LCA study, the limitations can be attributed to the design of LCA such as definition of system boundary, functional unit, data source; availability of commercial data, assumptions made in simplifying the LCA model.

Further studies are needed for the disputed environmental categories of acidification, eutrophication, photochemical, human and ecotocixity. Attention needs to be placed on water consumption and how it is influenced by different types of feedstock, irrigation and ethanol conversion technology. Finally, both water and human health issues need to feature more prominently next to those of climate change and fossil depletion concerns.

Acknowledgements

The research reported here was supported in full by the Biotechnology and Biological Sciences Research Council (BBSRC) Sustainable Bioenergy Centre (BSBEC), under the programme for 'Lignocellulosic Conversion to Ethanol' (LACE) [Grant Ref: BB/G01616X/1]. This is a large interdisciplinary programme and the views expressed in this paper are those of the authors alone, and do not necessarily reflect the views of the collaborators or the policies of the funding bodies. The authors would also like to thanks the anonymous reviewers for helping to make this a better paper.

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